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<b>(21) International Application Number:</b> PCT/US95/08358 <b>(22) International Filing Date:</b> 28 June 1995 (28.06.95)  <b>(30) Priority Data:</b> 08/267,603      28 June 1994 (28.06.94)      US  <b>(71) Applicant:</b> FOSTER-MILLER, INC. [US/US]; 350 Second Avenue, Waltham, MA 02254 (US).  <b>(72) Inventors:</b> BONIS, Laszlo, J.; 52 Haven Street, Dover, MA 02030 (US). SCHULER, Peter, S.; 45 LaSalle Avenue, Framingham, MA 02101 (US). ADUR, Ashok, M.; 106 Carter Terrace, Hackettstown, NJ 07840 (US).  <b>(74) Agents:</b> CONLIN, David, G. et al.; Dike, Bronstein, Roberts & Cushman, 130 Water Street, Boston, MA 02109 (US).		<b>(81) Designated States:</b> AU, CA, FI, JP, KR, MX, NO, NZ, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).  <b>Published</b> <i>With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>
<b>(54) Title:</b> COMPATIBILIZED LCP BLENDS  <b>(57) Abstract</b>  The present invention provides alloys having at least one thermotropic liquid crystalline polymer, polyethylene terephthate and at least one compatibilizer and methods of making such alloys.		

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Compatibilized LCP Blends

**FIELD OF THE INVENTION**

The present invention provides alloys comprising thermotropic liquid crystalline polymers (LCPs) and polyethylene terephthalate (PET) and at least one compatibilizer.

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**BACKGROUND**

High performance plastics are in widespread use in many industries and there is much interest in developing new plastics which are economical and recyclable, as well as high performance. The blending and alloying of existing polymers is a cost effective way to produce new high performance plastics which meet these criteria.

Polymer blends containing thermotropic LCPs have received increasing attention in the scientific and technical literature. The range of high performance thermoplastic flexible polymers which have been blended with TLCPs include polyimides, polyamides, poly(ethersulfone) (PES), poly(etherimide) (PEI), polyetherketone (PEEK), polycarbonate (PC), poly(ethylene terephthalate) (PET), poly(ethylene naphthalate), polyphenylene sulfide (PPS), and polyarylate.

Thermotropic LCPs are a relatively new class of high performance polymeric materials which combine the advantages of melt processability and outstanding mechanical properties. Because of their rigid backbone structure with flexible spacer groups, commercially available thermotropic LCPs have far higher tensile strength and flexural moduli than conventional polymers. However, thermotropic LCPs are in many cases difficult to process without specialized equipment and very costly as compared with conventional polymers when used alone.

Blending thermotropic LCPs with other polymers has been shown to improve processability of the other polymers, particularly LCPs based on wholly aromatic chain segments. Furthermore, blending with conventional thermoplastic polymers reduces costs, because less of the very costly LCP is used. Also, because thermotropic LCPs form an ordered phase in the melt

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(hence, the name thermotropic), they have shear viscosities far lower than other polymers and thus, have potential importance as a processing aid in mixtures with other polymers by reducing the melt viscosity of the mixture.

Thermotropic LCP in blends with PET have been reported to act as a "flow aid" at levels of 5-10% by reducing the melt viscosity. In U.S. Patent Nos. 4,386,174, 4,433,083, and 4,438,236, it is disclosed that blending a thermotropic LCP with other polymers such as PET changes the melt viscosity of PET. At 10% loading (LCP) the viscosity of PET is reduced to 25-50% of its original viscosity. O'Brien and Crosby (O'Brien, G.S. and Crosby, J.M., Proceedings of COMPALLOY '91 Conference, January 30-February 1, 1991, pp. 133-148) described LCP/PTFE blends to improve the flow of PTFE in the melt.

The use of thermotropic LCPs in blends to provide "reinforcement," especially where the LCP has a very rigid structure has been reported. XYDAR® (Poly(oxybenzoyl-co-bisphenyl terephthalate), Amoco, and VECTRA® Poly(oxybenzoyl-co-oxynaphthoyl), Hoechst-Celanese, are thermotropic LCPs which have been much studied as blend components. Crevecoeur, G. and Groeninckx, G., *Polymer Eng. Science*, 30, 532 (1990), reported that a thermotropic LCP can be used at 5-30% levels in polystyrene so that the LCP forms a disperse phase. In a 75:25 Polystyrene/VECTRA® A950 LCP blend at a draw ratio of 5, the LCP phase was reported as being slightly elongated. However at a draw ratio of 10 or more, the LCP phase was reported to show a well-developed microfibrillar morphology and to display a substantial increase in elastic modulus over a compression or injection-molded sample.

The use of LCPs in blends with thermoplastic polymers, e.g., PC and PET, to achieve improved mechanical properties over those of the thermoplastic polymer alone was reported in 1989. See, Bonis, L.J., "Multilayer Thermoplastics Advance Composites By Coextrusion", *The Polymer Processing Society Summer Meeting*, Amherst, Massachusetts, August 16-17, 1989, Paper 10F. See, also Williams, D.J., Proceedings of

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COMPALLOY '91 Conference, January 30 - February 1, 1991, pp. 393-408 which describes potential applications for thermotropic liquid crystal polyester blends.

Polymer molding compositions containing polycarbonates, thermoplastic polyester, and liquid crystalline polymers, wherein the liquid crystalline polymer is present as droplets or low aspect ratio particles, are disclosed in U.S. Patent No. 5,262,473. In the process disclosed in U.S. Patent No. 5,262,473, compatible blends of the polyester and polycarbonate may be used. Other blends are disclosed in, for example, U.S. Patent Nos. 5,070,157 and 5,156,785.

A blend is a physical mixture of two or more components which typically offers a compromise of properties and economies of the individual components. It is well known that the nature and properties of the interface of components in a blend frequently exert a limiting effect on the bulk properties of a multi-phase blend material. In fact, the physical and mechanical properties of a blend are very often inferior to the mathematical average of the properties of the original components. Blend components can be miscible or immiscible in their behavior toward each other.

Alloys are different from blends. Although they are also composed of two or more components, alloys exhibit strong intermolecular forces wherein intermolecular bonding between the components of the blend is provided by compatibilizers. This bonding in turn, creates new properties different from those of the original components and often exceeding those of the average of the original ingredients. The types of interaction or "chemical bonding" between the components can include, for example, one or more of the following mechanisms: ionic; covalent; molecular inter-penetration; hydrogen bonding; or associative.

Successful compatibilization by one or more of these interactions gives rise to interfacial adhesion to provide the formation of cohesive multi-phase compatibilized alloys with useful properties. To achieve compatibilization a number of strategies have emerged.

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In one approach, suitable block or graft copolymers are introduced to serve as macromolecular emulsifiers providing covalent bonds that traverse and fortify the blend interface. Block and graft copolymers may be generated *in-situ* through reactive extrusion and blending to generate a compatibilized blend.

In another approach, polymers having nucleophilic functional groups are interacted with compatibilizers containing hydrogen to form hydrogen bonding. Ionomers have also served as compatibilizers. In some cases, ionic or strong physicochemical interactions are generated across the interface, which in turn enhances compatibilization.

Compatibilization can also result from the addition of a similar functional group using the "like attract like" theory, such as the use of chlorinated polyethylene to compatibilizer polyvinyl chloride with polyethylene. This has been referred to as "associative" bonding.

Finally, compatibilization has even been demonstrated by the addition of a third immiscible phase component that exhibits relatively low interfacial tension with each of the primary blend components, i.e., those components intended to be compatibilized. The compatibilizing effects of the mutually miscible component may result from its presumed tendency to become enriched in the vicinity of the blend interface.

Alloying provides a tool to lower the cost of high performance resins while at the same time retaining many of the desirable properties and/or providing improved properties such as increased processability. The most successful alloying procedures result in a controlled and stable morphology with a singular thermodynamic profile. However, even when alloying is not "complete" in the multi-component system useful compositions can result.

At present, there is no known direct compatibility between LCPs and PET.

Accordingly, approaches to compatibilize LCPs with PET and, thereby, to provide LCP/PET alloys having properties which can be tailored to meet end-use specifications are being sought.

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**SUMMARY OF THE INVENTION**

The present invention provides alloys comprising a thermotropic LCP, PET, and at least one compatibilizer. In one preferred embodiment, two compatibilizers are present.

5 Preferred compatibilizers include copolyester elastomers; ethylene-unsaturated ester copolymers, such as ethylene-maleic anhydride copolymers; copolymers of ethylene and a carboxylic acid or acid derivative, such as ethylene-methyl acrylate copolymers; polyolefins or ethylene-unsaturated ester copolymers grafted with functional monomers,  
10 such as ethylene-methyl acrylate copolymers; copolymers of ethylene and a carboxylic acid or acid derivative, such as ethylene-methyl acrylate-maleic anhydride terpolymers; terpolymers of ethylene, unsaturated ester and a carboxylic acid or acid derivative, such as ethylene-methyl-methacrylic acid terpolymers; and acrylic elastomers, such as acrylic rubbers. A particularly  
15 preferred copolyester elastomer is HYTREL™ HTR-6108; ethylene-maleic anhydride copolymer is Polybond™3009; ethylene-methyl acrylate copolymer is SP 2205™; ethylene-methyl acrylate copolymer grafted with maleic anhydride is DS1328/60™; ethylene-methyl acrylate-maleic anhydride  
20 terpolymer is Lotader™ 2400; ethylene-methyl-methacrylic acid terpolymer is Escor™ ATX-320, Escor™ ATX-325 or Escor™ XV-11.04; and acrylic rubber is Vamac™ G1.

Preferred thermotropic LCPs include wholly or partially aromatic polyesters or copolyesters. A particularly preferred copolyester is  
25 XYDAR™ or VECTRA™. Other preferred thermotropic liquid crystal polymers include SUMIKOSUPER® and EKONOL™ (Sumitomo Chemical), DuPont HX™ and DuPont ZENITE™ (E.I. duPont de Nemours), RODRUN® (Unitika) and GRANLAR™ (Grandmont).

Preferred LCPs for use in the present invention include any such resins with a melt temperature in the range of 250 to 320°C. Particularly  
30 preferred LCPs have a melt temperature in the range of 250 to 280°C.

One preferred alloy in accordance with the present invention

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comprises PET, a wholly aromatic LCP copolyester and an ethylene-methyl acrylate-acrylic acid terpolymer compatibilizer, for example, Escor™ ATX-320, Escor™ ATX-325, or Escor™ XV-11.04.

5 Another preferred alloy comprises PET, a wholly aromatic LCP copolyester and an ethylene-maleic anhydride copolymer compatibilizer such as Polybond™ 3009.

Yet another preferred alloy in accordance with this invention comprises PET, a wholly aromatic LCP copolyester and an ethylene-methyl acrylate copolymer grafted with maleic anhydride compatibilizer, such as  
10 DS™ 1328/60, or a copolyester elastomer such as HYTREL™ HTR 6108.

Alloys comprising PET, LCP and at least two compatibilizers are particularly preferred in the practice of the present invention. The compatibilizers are preferably selected from a copolyester elastomer, ethylene-maleic anhydride copolymer, ethylene-methyl acrylate copolymer,  
15 ethylene-methyl acrylate copolymer, ethylene-methyl acrylate-maleic anhydride terpolymer, ethylene-methyl-methacrylic acid terpolymer or acrylic rubber.

Preferred two compatibilizer alloys include: PET, a wholly aromatic LCP copolyester, an ethylene-methyl acrylate-acrylic acid terpolymer and an  
20 ethylene-maleic anhydride copolymer compatibilizer. Exemplary ethylene-methyl acrylate-acrylic acid terpolymers include Escor™ ATX-320, Escor™ ATX-325, or Escor™ XV-11.04 and an exemplary ethylene-maleic anhydride copolymer is Polybond™ 3009.

In other preferred PET/LCP alloys, the LCP comprises a wholly  
25 aromatic copolyester and the compatibilizers are an ethylene-methyl acrylate copolymer and/or an ethylene-maleic anhydride copolymer. An exemplary ethylene-methyl acrylate copolymer is SP 2205™ and an exemplary ethylene-maleic anhydride copolymer is Polybond™ 3009.

Another preferred LCP/PET alloy of the present invention comprises  
30 a wholly aromatic LCP copolyester and an ethylene-methyl acrylate copolymer grafted with maleic anhydride and an ethylene-maleic anhydride



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copolymer compatibilizer. Yet another preferred alloy comprises PET, wholly aromatic LCP copolyester, and copolyester elastomer such as HYTREL™ HTR 6108 and an ethylene-maleic anhydride copolymer such as Polybond™ 3009.

5       The ethylene-methyl acrylate copolymer grafted with maleic anhydride, DS™ 1328/60, and the ethylene-maleic anhydride copolymer, Polybond™ 3009 are particularly preferred when the LCP is VECTRA™. Also preferred when the LCP is VECTRA™ is the compatibilizer, Polybond™ 3009 and a second compatibilizer, Escor™ ATX-320, Escor™  
10    ATX-325, DS1328/60™, Escor™ XV-11.04, or HYTREL™6108.

      The properties of the LCP and PET, as well as desired properties of the resulting alloy, are all taken into consideration in selecting suitable compatibilizers for use in the present invention. The properties of the PET/LCP alloys of the present invention are adjusted by adjusting the  
15    amount of compatibilizer and, in some preferred embodiments, by the manner in which the components are combined.

      Because the most expensive component in the alloys of the present invention typically is the LCP, in order to reduce costs it is preferable to keep the LCP content of the composition as low as possible while achieving  
20    the desired effect. Hence, in the present alloys the LCPs are used as the disperse phase, whereas PET is used as the predominant or bulk phase.

      When no compatibilization exists between PET and LCP, such as when no compatibilizer is present, the mechanical properties of the resulting blend are low. For example, in films extruded from blends comprising  
25    10% LCP / 90% PET (KODAR™ A-150) a machine direction (MD) tensile strength of only about 6,000 psi and MD tensile modulus of only about 300,000 psi are obtained. Furthermore, the oxygen barrier properties are poor, for example, around 35 to 40 cc/100in.<sup>2</sup> It was unexpectedly found that when PET/LCP alloys were formed by adding suitable compatibilizers  
30    in accordance with the teachings of the present invention, improved mechanical properties and/or lower gas permeation (barrier) numbers were

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obtained.

The present invention also provides methods of preparing the alloys described above. These methods include:

- i. LCP, PET and at least one compatibilizer are  
5 mixed and melt blended to form an alloy;
- ii. LCP, PET and a portion of the total  
compatibilizer to be used are mixed and melt  
blended, the remainder of the compatibilizer is  
added at a later time and further melt blended;
- 10 iii. LCP, PET and a first compatibilizer are mixed  
and melt blended. A second compatibilizer is  
added to the melt blend at a later time and  
further melt blended;
- iv. LCP and PET are mixed and melt blended and  
15 at least one compatibilizer is added at a later  
time to the melt blend and further melt  
blended;
- v. PET is melted under appropriate conditions in  
an extruder and at a later time LCP and at least  
20 one compatibilizer are added to the PET and  
further melt blended;
- vi. PET and a first compatibilizer are melt blended  
and at a later time LCP and a second  
compatibilizer are added to the melt blend and  
25 further mixed and melt blended;
- vii. PET and LCP are mixed and melt blended and  
two compatibilizers are added to the melt  
blended and further melt blended; and
- viii. PET, LCP and two compatibilizers are mixed  
30 and simultaneously melt blended.

**DETAILED DESCRIPTION OF THE INVENTION**

The LCP/PET alloys of the present invention are formed by use of at least one compatibilizer. In one preferred embodiment, two compatibilizers are used to form the alloys.

5           The alloys of the present invention comprise from about 0.5 to about 10 weight percent thermotropic liquid crystalline polymer, from about 40 to about 90 weight percent PET, and from about 1 to about 50 weight percent compatibilizer.

10           The liquid crystalline polymer is preferably present in amounts from about 5 to about 10 weight percent, PET is preferably present in amounts from about 70 to about 93 weight percent and one or more compatibilizers are present in amounts from about 2 to about 20 weight percent.

15           In a particularly preferred embodiment, the compositions of the present invention contain from about 9 to about 12 weight percent LCP, from about 78 to about 86 weight percent PET, and from about 5 to about 10 weight percent compatibilizer.

20           Suitable PET for use in the present invention is prepared by the reaction of either terephthalic acid or dimethyl terephthalate with ethylene glycol. Other methods of making suitable PET are known to one of ordinary skill in the art. Suitable PET is also available commercially from a number of vendors, including KODAR™ A150, KODAR™ 9921, and Eastman 13339 from Eastman Chemical and CPET™ from Goodyear. KODAR™ A150 is one preferred commercially available PET for use in the present invention.

25           Suitable thermotropic LCPs for use in the present invention include wholly and partially aromatic polyesters and co-polyesters such as those disclosed in U.S. Patent Nos. 3,991,014, 4,067,852, 4,083,829, 4,130,545, 4,161,470, 4,318,842, and 4,468,364. Preferred thermotropic LCPs for use in the present invention include wholly aromatic co-polyester. Vectra™  
30           A950, sold by Celanese Research Corporation, Summit, New Jersey is one such wholly aromatic copolyester. This polymer has been reported to

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consist essentially of about 25-27 percent of 6-oxy-2-naphthoyl moieties and about 73-75 percent of p-oxybenzoyl moieties, as described in example 4 of U.S. Patent No. 4,468,364 and in G. W. Calundann et al., "Anisotropic Polymers, Their Synthesis and Properties", reprinted from Proceedings of the Robert A. Welch Conferences on Chemical Research, XXVI Synthetic Polymers, November 15-17, 1982, Houston, Texas, pp. 247-291 (see especially pp. 263-265).

Preferred thermotropic LCPs include wholly or partially aromatic polyesters or copolyesters. A particularly preferred copolyester is XYDAR™ or VECTRA™. Other preferred thermotropic liquid crystal polymers include SUMIKOSUPER® and EKONOL™ (Sumitomo Chemical), DuPont HX™ and DuPont ZENITE™ (E.I. duPont de Nemours), RODRUN® (Unitika) and GRANLAR™ (Grandmont).

In formulating the composition of the alloys of the present invention a number of variables including, the properties of the polymers to be blended, properties of the compatibilizers, and the amount and ratio of the components, are taken into consideration. These variables are tailored and optimized in accordance with the present teachings to provide alloys to meet a particular end use specification. For example, if high gas barrier properties are desired, then polymers having high individual gas barrier properties are preferably selected.

The amount of compatibilizer is adjusted to provide intermolecular bonding among the components of the alloy to enhance properties and at the same time, to avoid the formation of a quasi- or pseudo-cross linked network which is not readily processable.

The compatibilizers for use in the present invention are either miscible with each of the LCP and the PET through, e.g., covalent, ionic, molecular inter-penetration, hydrogen bonding or associative interactions as mentioned above, or have interactive miscibility when the LCP and PET are present in a common phase. In other words, the functional groups of the compatibilizer, LCP, and PET for use in the alloys are also chemically

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compatible. For example, if the LCP to be alloyed with PET has an aliphatic type of polyester functionality, such as acrylate or methacrylate, or an aromatic functionality, such as a benzoate or phthalate ester linkage, then preferred compatibilizers will have a functionality, such as a polyester functional group or a maleic anhydride functional group, that is capable of reacting with the polyester group.

Compatibilizers for use in the present invention are also processable in the melting and processing range of PET and the LCP and exhibit temperature stability at the intended processing temperature. By temperature stability is meant that a component of the alloy essentially retains its chemical functionality and, hence, its interfacial interaction with the other components of the alloy with which it interacts. If one of the components were not thermally stable, it is possible that the compatibilization achieved could fail on subsequent processing.

Preferred alloys of the present invention comprise at least one thermotropic LCP, PET, and at least one compatibilizer. Particularly preferred embodiments include two or more compatibilizers, wherein at least one compatibilizer interacts with the LCP and at least one interacts with the PET. The ratios of compatibilizers to each other and in the total composition are adjusted to achieve alloys having the desired properties as is shown in the examples which follow.

The following compatibilizers are particularly preferred in the practice of the present invention wherein components of the alloy comprise PET and wholly aromatic esters and copolyesters liquid crystal polymers, such as VECTRA™ and XYDAR™:

- i. Copolyester elastomers such as HYTREL™  
HTR-6108 from DuPont;
- ii. Ethylene-maleic anhydride copolymers (HDPE grafted with maleic anhydride), such as  
Polybond™ 3009 from BP Chemicals;
- iii. Ethylene-methyl acrylate copolymers, such as

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SP 2205 from Chevron;

- iv. Ethylene-methyl acrylate copolymers grafted with maleic anhydride, such as DS 1328/60 from Chevron;
- 5 v. Ethylene-methyl acrylate-maleic anhydride terpolymers, such as Lotader™ 2400 from Atochem;
- vi. Ethylene-methyl-methacrylic acid terpolymers (ethylene-methyl acrylate-acrylic acid terpolymers) such as Escor™ ATX-320, Escor™ ATX-325, and Escor™ XV-11.04; and
- 10 vii. Acrylic rubber such as VAMAC™ from DuPont.

15 The alloys of the present invention can be extruded to form various articles of manufacture such as films and tubes useful, e.g., in food packaging, electronic circuit substrates and structural applications. The films can be thermoformed to provide, e.g., trays, blow molded to, e.g., form containers, and otherwise processed by known methods. In some

20 embodiments, articles of manufacture comprising the alloys of the present invention are provided with a thin coating of, e.g., glass, metal or another polymer both to protect the article and to provide suitable means to affix labels and the like.

To illustrate the improved properties of the alloys of the present

25 invention, various alloys were prepared as taught herein and extruded to form films having improved tensile strength, tensile modulus and/or oxygen barrier properties over films extruded from PET or LCP and PET blends without compatibilizers. In some films, tensile strength was increased by up to more than 2 times and tensile modulus was increased up to more than 3

30 times over that of the blend without compatibilizer. In many instances, values above 10,000 psi and tensile strength and/or above 500,000 psi and

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tensile modulus were obtained.

Films extruded from alloys comprising PET, and an anhydride-grafted ethylene-methyl acrylate copolymer, thermotropic LCP wholly aromatic copolyester and an ethylene-maleic anhydride copolymer showed  
5 great improved mechanical properties.

Films extruded from three component alloys comprising PET, wholly aromatic copolyester and an ethylene-methyl-metha acrylic acid terpolymer, e.g., Escor™ ATX-320 or -325, had superior mechanical properties. Also, three component blends comprising PET, wholly aromatic copolyester and a  
10 copolyester elastomer such as HYTREL™ HTR-6108 were extruded to produce films having superior mechanical properties.

A number of films extruded from the alloys of the present invention yielded low oxygen permeation values, ranging from about 18 to 30, well below the 36 to 40 cc/100in.<sup>2</sup> expected for PET.

Films extruded from alloys comprising PET, a wholly aromatic copolyester LCP and a copolyester elastomer such as HYTREL™ 6108 had  
15 excellent barrier properties. Also, films extruded from alloys comprising PET, a copolyester elastomer, such as HYTREL™ 6108, a wholly aromatic copolyester, and ethylene-maleic anhydride copolymer, such as Polybond™  
20 3009 had excellent oxygen barrier properties, e.g., from about 21 to 23 cc/100in<sup>2</sup>.

Optional components well known to the skilled artisan may be added to alloys of the present invention provided that they do not interfere with formation or with the desired final properties of an alloy. Such additives  
25 includes fillers and pigments, lubricants, mold release agents, plastizers, ultraviolet stabilizers and so forth.

In the methods of the present invention, compatibilizers are used either alone or in various combinations with LCP and PET to achieve the desired results. They are also used in single step and sequential  
30 compatibilization methods as described below.

The following methods have been found to provide alloys having

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improved properties which can be used, e.g., to provide films having improved properties over films of LCP and PET blends. These methods include:

- 5           i.       LCP, PET and at least one compatibilizer are mixed and melt blended to form an alloy;
- ii.       LCP, PET and a portion of the total compatibilizer to be used are mixed and melt blended, the remainder of the  
10           compatibilizer is added at a later time and further melt blended;
- iii.       LCP, PET and a first compatibilizer are mixed and melt blended. A second  
                      compatibilizer is added to the melt blend  
15           at a later time and further melt blended;
- iv.       LCP and PET are mixed and melt blended and at least one compatibilizer is  
                      added at a later time to the melt blend  
                      and further melt blended;
- 20           v.       PET is melted under appropriate conditions in an extruder and at a later  
                      time LCP and at least one compatibilizer are added to the PET and further melt  
                      blended;
- 25           vi.       PET and a first compatibilizer are melt blended and at a later time LCP and a  
                      second compatibilizer are added to the melt blend and further mixed and melt  
                      blended;
- 30           vii.       PET and LCP are mixed and melt blended and two compatibilizers are



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added to the melt blended and further  
melt blended; and

- viii. PET, LCP and two compatibilizers are  
mixed and simultaneously melt blended.

5

By controlling the order in which the components of the alloys are mixed  
and melt blended the properties of the alloy are controlled to enable the  
production of articles of manufacture, e.g., films, which have improved  
properties over the properties of a similar article of manufacture composed  
solely of PET or of LCP and PET.

10

In the production of films from the alloys described above, the melt  
blend is extruded, e.g., through a slot die, a circular, counter-rotating die,  
or a circular rotating trimodal die.

15

In alloys containing two compatibilizers, sequential compatibilization  
according to methods (iii) and (vi) above are preferred preparation methods.  
It was unexpectedly discovered that these unique methods of combining two  
or more compatibilizers, provided alloys having improved properties.  
While not wishing to be bound by theory, it is believed that in this novel  
process, two compatibilizers interact sequentially to provide the desired  
compatibilization and in some cases also interact with each other. In the  
case of PET-LCP blends of the present invention, the interaction is between  
the PET and a first compatibilizer, and the LCP interacts with a second  
compatibilizer. The products of these two interactions, then sequentially  
react with one another to form an alloy.

20

25

The methods of the present invention provide a great deal of  
flexibility to achieve the desired compatibilization through the wide array of  
possibilities for the compatibilizers to interact with the major components of  
the alloy, which is the object of the compatibilization. The method of the  
present invention is an innovative yet efficient way to achieve the desired  
end results.

30

In one preferred embodiment of the present invention, Chevron DS

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1328/60, an anhydride-grafted ethylene-methyl acrylate copolymer, was melt blended with PET and then Polybond™ 3009, an ethylene-maleic anhydride copolymer, and VECTRA™ A950 were added to the mixture and further melt blended to produce alloys which were extruded to produce films having greatly improved mechanical properties. For example, a tensile strength of 14,800 psi was obtained in one film extruded from an alloy made by feeding 5% of the Chevron DS 1328/60 in the hopper with the PET, and then by feeding 2% Polybond™ 3009 with the LCP VECTRA™ A950 into the vent feed port.

10 In another preferred embodiment, Escor™ ATX-325, an ethylene-methyl-methacrylic acid terpolymer, was melt blended with PET and then Polybond™ 3009 and Vectra™ A950 were added to the mixture and further melt blended. A tensile modulus value of 1.09 million psi was obtained in a film extruded from an alloy made by feeding 5% of Exxon ATX™ 320 in the hopper with the PET, and then feeding 2% Polybond™ 3009 with the LCP VECTRA™ A950 into the vent feed port.

Accordingly, it can be seen that films produced from the alloys of the present invention have surprisingly improved properties over films of LCP and PET blends or of PET alone.

20 Conventional extrusion equipment was used to produce the alloys of the present invention and to extrude films from these alloys. Mixing and melt blending of components to form the alloys of the present invention is carried out using conventional single or double screw extruders. It is preferred that the extruder system has not less than 25/1 L/D ratio and not smaller than 2". Extrusion conditions such as processing temperatures, rotation speed of the screw, feed rate and through put were optimized for the particular alloy by taking into consideration the properties of the polymers being melt blended to form the alloy, including resulting viscosity of the melt blend. Typically, higher shear screw configurations were found to give better dispersions of the LCP and better compatibilization resulting in alloys that could be used to produce films having improved properties.

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Typical temperatures employed for the processing were 525 to 580°F. The rotation rate of the screw was, typically between 50 to 200 rpm.

5 The alloy components are appropriately conditioned, e.g., dried and then fed to the extruder using conventional methodologies. For example, the components can be melt blended and extruded to form pellets. The pellets can then be extruded or injection molded to form the desired article of manufacture. Alternatively, the dry components can be blended in a hopper and extruded, e.g., to a film directly.

10 The masterblending or masterbatching technique in which typically, a blend of two components is processed into pellets to form the "masterblend" can also be used. The masterblend can be run through an extruder a second time with additional components added in accordance with the teachings of the present invention. This is a convenient method of manufacture, because an inventory of masterblend material can be made and then combined with  
15 different components as desired. One advantage to the masterblending process is that small and very controlled amounts of additional components can be added to the masterblend. For example, if the masterbatch has 10% LCP, the masterbatch can be passed through the extruder again with, for example, 10% of the masterbatch and 90% of the other polymers, providing  
20 a masterbatch that is 1% in LCP.

Through masterbatching, controlled low concentration of a component in the alloy can be obtained; and additional mixing and shearing through multi-passes in the extruder can be achieved, if desired.

25 The present invention will be further illustrated with reference to the following example which is intended to aid in the understanding of the present invention, but which is not to be construed as a limitation thereof.

#### EXAMPLE

30 The alloy components should be appropriately treated, e.g., dried, before processing as would be readily apparent to the skilled artisan.

The work described in the following example was carried out using a

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conventional 25 mm or 40 mm co-rotating, non-intermeshing twin screw extruder manufactured by Berstorff Corporation. Mixing and kneading elements for the screw configuration were varied according to conventional wisdom to achieve the desired degree of mixing.

5           Films were extruded from a slot die, approximately 8 inches wide with die gap of approximately 0.010 to 0.020 inches. Also, a counter-rotating die or circular trimordal die (see, U.S. Patents 4,975,312 and 5,288,529) can be used to extrude films comprising one or more alloys of the present invention. Since the degree of uniaxial orientation produced in  
10       the extruded film has an impact on the properties, films having similar extrusion conditions were compared in the work discussed below.

A universal testing machine was used for testing the tensile properties based on ASTM standard tests, e.g., ASTM #0882.

The LCP used was Vectra™ A-950 from Hoechst-Celanese  
15       Corporation. PET purchased from Eastman Chemicals under the tradename KODAR™ A150 was used in the following work. The compatibilizers used included: Hytrel™ HTR-6108 from DuPont (Copolyester elastomer); Polybond™ 3009 from BP Chemicals (HDPE grafted with maleic  
20       anhydride); SP 2205 from Chevron (ethylene-methyl acrylate copolymer); DS 1328/60 from Chevron (ethylene-methyl acrylate copolymer grafted with maleic anhydride); Lotader™ 2400 from Atochem (ethylene-methyl acrylate-maleic anhydride terpolymer); Escor™ ATX-320 from Exxon Chemicals (ethylene-methyl acrylate acrylic acid terpolymer); Escor™ ATX-325 from  
25       Exxon Chemicals (ethylene-methyl acrylate-acrylic acid terpolymer); Escor™ XV-11.04 from Exxon Chemicals (ethylene-methyl acrylate-acrylic acid terpolymer); and Vamac™ G1 Acrylic rubber from DuPont.

The run conditions and results are shown in Tables I-VII. In the Tables, the PET used is indicated as "A150" or "9921" the VECTRA™ LCP is indicated as "A950." "Ten Yld St." indicates Tensile Yield Strength; and  
30       "Ten. Mod." indicates Tensile Modulus Values. In the Tables, compatibilizers listed are identified as follows: HYTREL™ HTR-6108 as

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Hytre 6108; Polybond™ 3009 as "BP3009"; SP 2205™ as "SP2205"; DS 1328/60 as "Chev DS"; Lotader™ 2400 as "Lotader 2400"; Escor™ ATX-320, ATX-325 and XV11.04 as "ATX320", "ATX325" and "XV11.04" respectively.

5

TABLE A

10 LCP, PET and at least one compatibilizer are mixed and melt blended to form an alloy

RUN #	HOPPER FEED	VENT FEED	TEN YLD ST MD(Kpsi)	TEN MOD MD(Kpsi)	OX- BARRIER
15 6299-7	85.7%A150+9.5%A950+4.8%ATX320	-	10.6	800	27.2
6299-9	85.7%A150+9.5%A950+4.8%ATX325	-	9.7	810	-
6299-10	85.7%A150+9.5%A950+4.8%Chevron	-	9.1	570	25.3

20

TABLE B

25 LCP, PET and a portion of the total compatibilizer to be used are mixed and melt blended, the remainder of the compatibilizer is added at a later time and further melt blended.

RUN #	HOPPER FEED	VENT FEED	TEN YLD ST MD(Kpsi)	TEN MOD MD(Kpsi)	OX- BARRIER
30 6249-3	85.7%A150+9.5%A950+2.4%ATX325	2.4%ATX325	10.0	421	25.5
6249-7	85.7%A150+9.5%A950+2.4%BP3009	2.4%BP3009	12.9	460	26.8

35

- 20 -

TABLE C

LCP, PET and a first compatibilizer are mixed and melt blended. A second compatibilizer is added to the melt blend at a later time and further melt blended.

RUN #	HOPPER FEED	VENT FEED	TEN YLD ST MD(Kpsi)	TEN MOD MD(Kpsi)	OX- BARRIER
6249-8	85.7%A150+9.5%A950+2.4%ATX325	2.4%Lotader2400	7.2	233	-

TABLE D

LCP and PET are mixed and melt blended and at least one compatibilizer is added at a later time to the melt blend and further melt blended.

RUN #	HOPPER FEED	VENT FEED	TEN YLD ST MD(Kpsi)	TEN MOD MD(Kpsi)	OX- BARRIER
6299-14	85.7%A150+9.5%A950	7.8%ATX320	9.1	530	26.6

TABLE E

PET is melted under appropriate conditions in an extruder and at a later time LCP and at least one compatibilizer are added to the PET and further melt blended.

RUN #	HOPPER FEED	VENT FEED	TEN YLD ST MD(Kpsi)	TEN MOD MD(Kpsi)	OX- BARRIER
6309-12	85.7%A150	9.7%A950+4.8%ATX325	7.1	460	-

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TABLE F

PET and a first compatibilizer are melt blended and at a later time LCP and a second compatibilizer are added to the melt blend and further mixed and melt blended.

RUN #	HOPPER FEED	VENT FEED	TEN YLD ST MD(Kpsi)	TEN MOD MD(Kpsi)	OX- BARRIER
7019-7	89%A150+4.7%ATX320	9.3%A950+2%BP3009	12.7	810	29.6
7069-8	89%A150+4.7%ChevronDS	9.3%A950+2%BP3009	11.2	960	25.7
7069-9	89%A150+4.7%XV11.04	9.3%A950+2%BP3009	9.6	740	-
7069-10	89%A150+4.7%Hytre6108	9.3%A950+2%BP3009	9.3	850	20.3
7079-1	89%A150+4.7%ChevronDS	9.3%A950+2%BP3009	14.8	840	29.8

TABLE G

PET & LCP are mixed and melt blended and two compatibilizers are added to the melt blended and further melt blended.

RUN #	HOPPER FEED	VENT FEED	TEN YLD ST MD(Kpsi)	TEN MOD MD(Kpsi)	OX- BARRIER
7069-6	85.7%A150+9.5%A950	2.4%Hytre6108+2.4%BP3009	6.2	420	26.3

TABLE H

PET, LCP and two compatibilizers are mixed and simultaneously melt blended in an extruder and extruded through a slot die.

RUN #	HOPPER FEED	TEN YLD ST MD(Kpsi)	TEN MOD MD(Kpsi)	OX- BARRIER
7169-3	80%A150+10%A950+7.5%Hytre6108+2.5%BP3009	7.9	370	25.3

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5

TABLE J

PET, LCP and two compatibilizers are mixed and simultaneously melt blended and extruded through a circular, counter-rotating die.

10

RUN #	HOPPER FEED	VENT FEED	TEN YLD ST MD(Kpsi)	TEN MOD MD(Kpsi)	OX-BARRIER
1199-3	83%A150+10%A950+5%ATX320+2%BP3009	-	6.2	440	-
1199-4	81%A150+10%A950+5%SP2205+4%BP3009	-	5.9	370	-
1199-5	81%A150+10%A950+5%SP2205+4%Hytre16108	-	7.2	440	-

15

20

TABLE K

PET and a first compatibilizer are melt blended and, at a later time, LCP and a second compatibilizer are added to the melt and further mixed and melt blended and extruded using a circular rotating trimodal die.

25

RUN #	HOPPER FEED	VENT FEED	TEN YLD ST MD(Kpsi)	TEN MOD MD(Kpsi)	OX-BARRIER
3249-2	83.6%A150+4.4%ATX320	10%A950+2%BP3009	9.0	290	-
3249-5	83.6%A150+4.4%ChevronDS	10%A950+2%BP3009	6.5	220	-

30



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TABLE L - Control PET

RUN #	HOPPER FEED	VENT FEED	TEN YLD ST MD(Kpsi)	TEN MOD MD(Kpsi)	OX- BARRIER
6249-1	100%A150	-	5.5	162	-
5269-0	100%A150	-	5.4	190	-
5119-0	100%A150	-	5.8	169	-
4239-1	100%A150	-	6.2	176	31.5
6299-1	100%A150	-	5.2	320	-
7069-1	100%A150	-	5.3	320	29.9
3189-1	100%A150	-	6.2	200	-

TABLE M - Control PET &amp; LCP, No Compatibilizer

RUN #	HOPPER FEED	VENT FEED	TEN YLD ST MD(Kpsi)	TEN MOD MD(Kpsi)	OX- BARRIER
3189-2	90%A150+10%A950	-	6.9	290	-
3189-5	90%A150+10%A950	-	5.5	210	22.7
3189-9	90%A150+10%A950	-	5.8	231	-
6249-2	90%A150+10%A950	-	7.4	251	-
7069-2	90%A150+10%A950	-	6.1	420	24.9
7019-5	90%A150+10%A950	-	6.1	440	-

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TABLE N - Masterbatch

RUN #	HOPPER FEED	VENT FEED	TEN YLD ST MD (Kpai)	TEN MOD MD (Kpai)	OX- BARRIER
8129-1	83%A150+5%Hytrel6108	10%A950+2%BP3009	8.0	550	20.6
8129-5	78%A150+10%Hytrel6108	10%A950+2%BP3009	7.8	230	19.6
8129-10	83%A150+5%Hytrel6108	10%A950+2%BP3009	5.6	430	22.1
8129-16	78%A150+10%Hytrel6108	10%A950+2%Hytrel6108	5.9	420	18.5
8129-17	78%A150+10%Hytrel6108	10%A950+2%Hytrel6108	5.8	430	18.4
8129-18	78%A150+10%Hytrel6108	10%A950+2%BP3009	7.4	600	19.5
8129-19	78%A150+10%Hytrel6108	10%A950+2%BP3009	7.6	710	18.0

The present invention has been described in detail including the preferred embodiments thereof. However, it would be appreciated that those skilled in the art, upon consideration of the present disclosure, may make modifications and/or improvements on this invention and still be within the scope and spirit of this invention as set forth in the following claims.

- 25 -

What is claimed is:

1. An alloy comprising at least one thermotropic liquid crystalline polymer (LCP), polyethylene terephthate (PET) and at least one compatibilizer.  
5
2. An alloy in accordance with claim 1, wherein the compatibilizer is a copolyester elastomer; an ethylene-unsaturated ester copolymer; a copolymer of ethylene and a carboxylic acid or acid derivative; a polyolefin or ethylene-unsaturated ester copolymer grafted with at least one functional monomer; a copolymer of ethylene and a carboxylic acid or acid derivative; a terpolymer of ethylene, unsaturated ester and a carboxylic acid or acid derivative; and an acrylic elastomer.  
10
3. An alloy in accordance with claim 2, wherein the copolyester elastomer is HYTREL™ HTR-6108; the ethylene-unsaturated ester copolymer is an ethylene-maleic anhydride copolymer; the copolymer of ethylene and a carboxylic acid or acid derivative is an ethylene-methyl acrylate copolymer; the polyolefin or ethylene-unsaturated ester copolymer grafted with at least one functional monomer is an ethylene-methyl acrylate copolymer; the copolymer of ethylene and a carboxylic acid or acid derivative is an ethylene-methyl acrylate-maleic anhydride terpolymer; the terpolymer of ethylene, unsaturated ester and a carboxylic acid or acid derivative is an ethylene-methyl-methacrylic acid terpolymer; and the acrylic elastomer is an acrylic rubber.  
15  
20  
25
4. An alloy in accordance with claim 3, wherein the ethylene-maleic anhydride copolymer is Polybond™3009; the ethylene-methyl acrylate copolymer is SP 2205™; the ethylene-methyl acrylate copolymer grafted with maleic anhydride is DS1328/60™; the ethylene-methyl acrylate-maleic anhydride terpolymer is Lotader™ 2400; the ethylene-methyl-methacrylic  
30

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acid terpolymer is Escor™ ATX-320, Escor™ ATX-325 or Escor™ XV-11.04; and the acrylic rubber is Vamac™ G1.

5           5.           An alloy in accordance with claim 1, wherein the thermotropic LCP is a wholly or partially aromatic polyester or copolyester.

6.           An alloy in accordance with claim 5, wherein the copolyester is XYDAR™ or VECTRA™.

10           7.           An alloy in accordance with claim 1, wherein the LCP is a wholly aromatic copolyester and the compatibilizer is an ethylene-methyl acrylate-acrylic acid terpolymer.

15           8.           An alloy in accordance with claim 7, wherein the ethylene-methyl acrylate-acrylic acid terpolymer is Escor™ ATX-320, Escor™ ATX-325, or Escor™ XV11.04.

20           9.           An alloy in accordance with claim 1, wherein the LCP is a wholly aromatic copolyester and the compatibilizer is an ethylene-maleic anhydride copolymer.

10.           An alloy in accordance with claim 9, wherein the ethylene-maleic anhydride copolymer is Polybond™ 3009.

25           11.           An alloy in accordance with claim 1, wherein the LCP is a wholly aromatic copolyester and the compatibilizer is an ethylene-methyl acrylate copolymer grafted with maleic anhydride.

30           12.           An alloy in accordance with claim 11, wherein the ethylene-methyl acrylate copolymer grafted with maleic anhydride is DS™ 1328/60.

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13. An alloy in accordance with claim 1, wherein the LCP is wholly aromatic copolyester and the compatibilizer is a copolyester elastomer.

5 14. An alloy in accordance with claim 13, wherein the copolyester elastomer is HYTREL™ HTR 6108.

15. An alloy in accordance with claim 1, wherein at least two compatibilizers are present.

10

16. An alloy in accordance with claim 15, wherein the compatibilizers are selected from a copolyester elastomer, ethylene-maleic anhydride copolymer, ethylene-methyl acrylate copolymer, ethylene-methyl acrylate copolymer grafted with maleic anhydride, ethylene-methyl acrylate-  
15 maleic anhydride terpolymer, ethylene-methyl-methacrylic acid terpolymer or acrylic rubber.

17. An alloy in accordance with claim 15, wherein the LCP is a wholly aromatic copolyester and the compatibilizers are an ethylene-methyl  
20 acrylate-acrylic acid terpolymer and an ethylene-maleic anhydride copolymer.

18. An alloy in accordance with claim 17, wherein the ethylene-methyl acrylate-acrylic acid terpolymer is Escor™ ATX-320,  
25 Escor™ ATX-325, or Escor™ XV11.04 and the ethylene-maleic anhydride copolymer is Polybond™ 3009.

19. An alloy in accordance with claim 15, wherein the LCP is a wholly aromatic copolyester and the compatibilizers are an ethylene-  
30 methyl acrylate copolymer and an ethylene-maleic anhydride copolymer.

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20. An alloy in accordance with claim 19, wherein the ethylene-methyl acrylate copolymer is SP 2205™ and the ethylene-maleic anhydride copolymer is Polybond™ 3009.

5 21. An alloy in accordance with claim 15, wherein the LCP is a wholly aromatic copolyester and the compatibilizers are an ethylene-methyl acrylate copolymer grafted with maleic anhydride and an ethylene-maleic anhydride copolymer.

10 22. An alloy in accordance with claim 21, wherein the ethylene-methyl acrylate copolymer grafted with maleic anhydride is DS™ 1328/60 and the ethylene-maleic anhydride copolymer is Polybond™ 3009.

15 23. An alloy in accordance with claim 15, wherein the LCP is a wholly aromatic copolyester and the compatibilizers are a copolyester elastomer and an ethylene-maleic anhydride copolymer.

20 24. An alloy in accordance with claim 23, wherein the copolyester elastomer is HYTREL™ HTR 6108 and the ethylene-maleic anhydride copolymer is Polybond™ 3009.

25 25. An alloy in accordance with claim 6 having at least two compatibilizers, wherein one compatibilizer is Polybond™ 3009 and one compatibilizer is Escor™ ATX-320, Escor™ ATX-325, DS1328/60™, Escor™ XV-11.04, or HYTREL™ 6108.

30 26. A method of producing an alloy comprising at least one thermotropic LCP, PET and at least one compatibilizer, wherein the method comprises melt blending the LCP, PET and compatibilizer to form an alloy.

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27. A method in accordance with claim 26, wherein the method comprises melt blending LCP, PET and a portion of a compatibilizer to form a first melt blend, and adding the remainder of the compatibilizer to the first melt blend and further melt blending to form an alloy.

28. A method of producing an alloy comprising at least one thermotropic LCP, PET and a first and second compatibilizer, wherein the method comprises melt blending PET and the first compatibilizer to form a first melt blend, adding LCP and second compatibilizer to the first melt blend, and further melt blending to form an alloy.

29. A method in accordance with claim 28, wherein the method comprises melt blending LCP, PET and the first compatibilizer to form a melt blend, adding the second compatibilizer to the melt blend, and further melt blending to form an alloy.

30. An article of manufacture comprising an alloy according to claims 1 or 2.

31. An article of manufacture according to claim 30, wherein the article comprises a film, a sheet, a tube, or a container.

32. An article of manufacture according to claim 30, wherein the article is provided with a coating comprising glass, metal or polymer.

## INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US95/08358

## A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) : C08L 67/02

US CL : 428/36.9, 36.92, 430, 458, 480; 525/68, 133, 444

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 428/36.9, 36.92, 430, 458, 480; 525/68, 133, 444

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US, A, 4,845,169 (ABU-ISA ET AL) 04 JULY 1989, See entire document.	1-6, 13-16, 23-32
X --- Y	EP, A, 0,369,734 (AMOCO CORPORATION) 23 MAY 1990. See entire document.	1-8, 15, 16 26-32 ----- 13, 14, 17, 18, 23-25
X --- Y	WO, A, 93/24574 (NESTE OY) 09 December 1993. See entire document.	1-12, 15-22 25-32 ----- 13, 14, 23, 24



Further documents are listed in the continuation of Box C.



See patent family annex.

* Special categories of cited documents:	* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
*A* document defining the general state of the art which is not considered to be of particular relevance	*X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
*E* earlier document published on or after the international filing date	*Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
*L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	*Z* document member of the same patent family
*O* document referring to an oral disclosure, use, exhibition or other means	
*P* document published prior to the international filing date but later than the priority date claimed	

Date of the actual completion of the international search

26 SEPTEMBER 1995

Date of mailing of the international search report

15 NOV 1995

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# INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US95/08358

## Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This international report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:  
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
3. ☐ Claims Nos.:  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

## Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

Please See Extra Sheet.

1. ☒ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.  
☐ No protest accompanied the payment of additional search fees.

# INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US95/08358

## BOX II. OBSERVATIONS WHERE UNITY OF INVENTION WAS LACKING

This ISA found multiple inventions as follows:

- I. Claims 1-12, 15-22 and 25-32 drawn to compositions comprising LCP, PET and an ethylene/acid copolymer classified in class 525, subclass 133.
- II. Claims 1-3, 5, 6, 13-14, 26, 27 and 30-32 drawn to compositions comprising LCP, PET and a copolyester elastomer classified in class 525, subclass 444.
- III. Claims 1-6, 15, 16, 23-32 drawn to compositions comprising LCP, PET, an ethylene/acid copolymer and a copolyester elastomer classified in class 525, subclass 133.

The listed inventions lack unity of invention under PCT Rule 13.1 to 13.3 because there is no special technical feature which links the diverse compositions of Groups I, II and III. There is no special technical feature because the compatibilizers do not belong to a recognized class of compounds.